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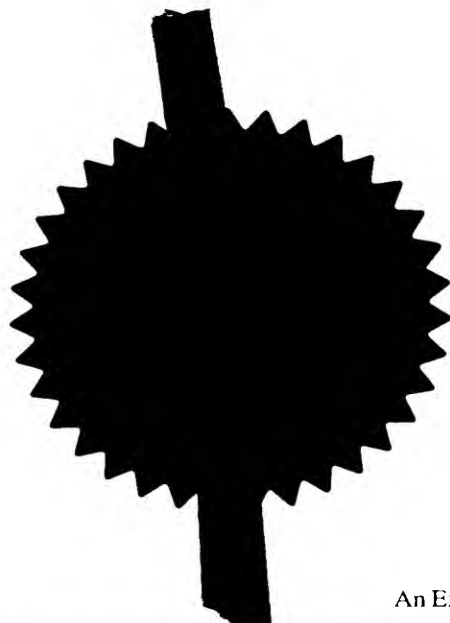
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Request for grant of a patent

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1	Your reference	MRH/P15693		
2	Patent application number	9919606.5		
3	Full name, address and postcode of the applicant	British Nuclear Fuels plc Risley Warrington Cheshire WA3 6AS 350108001		
	Patents ADP number			
	State of incorporation	UK		
4	Title of the invention	PROCESS FOR RECYCLING IONIC LIQUIDS		
5	Name of agent	Harrison Goddard Foote		
	Address for service	Belmont House 20 Wood Lane Headingley Leeds LS6 2AE 14571001 0113 2258350		
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6	Priority applications	Country	Priority App No	Date of Filing

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7	Parent application (eg Divisional)	Earlier Application No	Date of Filing
8	Statement of Inventorship Needed?		
9	Number of sheets for any of the following (not counting copies of same document) Continuation sheets of this form Description Claims Abstract Drawings	<div>4</div> <div>3</div>	
10	Number of other documents attached Priority documents Translations of priority documents P7/77 P9/77 P10/77 Other documents		
11	I/We request the grant of a patent on the basis of this application. Signature <u>Michael R Harrison</u> Date 19 Aug 1999		
12	Name and daytime telephone number of person to contact in the United Kingdom	Michael R Harrison <u>+44</u> 113 2258350	

DUPLICATE

PROCESS FOR RECYCLING IONIC LIQUIDS

FIELD OF THE INVENTION

5 This invention relates to processes for treating spent ionic liquids. Reference will be made hereinafter to ionic liquids which have been used in the reprocessing of nuclear fuels but it should be understood that the invention has application in all fields of ionic liquids technology, including applications outside the nuclear industry.

BACKGROUND OF THE INVENTION

10 Ionic liquids free of molecular solvents were first disclosed by Hurley and Wier in a series of US patents (24446331, 2446339, 2446350). In general terms an ionic liquid is a salt, a mixtures of salts, or a mixture of components which produce a salt or salts, which melts below or just above room temperature. (As used herein, the term "salt" means an entity comprising entirely of cationic and anionic species). Such liquids
15 are known as "ionic liquids" although this term is sometimes used for salts which melt at relatively high temperatures. In this specification, the term "ionic liquid" refers to a salt which melts at a temperature of up to 100°C.

20 Co-pending patent application PCT/GB99/00246 discloses a method for reprocessing spent nuclear fuel which comprises dissolving the spent fuel or constituent parts of the spent fuel in an ionic liquid to substantially separate fissile material from other components of irradiated fuel. Also disclosed is the subsequent treatment of the resulting ionic liquor, either by solvent extraction or electrochemical treatment to recover the dissolved uranium or plutonium.

25 In addition to fissile material spent ionic liquid also contains fission products which have been dissolved together with the uranium. Considerable effort has been focused on the development of a cost effective route for the removal of these contaminants, subsequent to the separation of the uranium from the mixture, in a form suitable for
30 conversion into a stable waste form.

Contaminant removal may be achieved by any one or a combination of a number of different routes. Selected fission products such as plutonium can be electrochemically extracted through the application of a suitable potential. The disadvantage of this is that electrolysis is a costly process, especially when
5 performed on an industrial scale.

Fission products may alternatively or additionally be extracted from the ionic liquid by the addition of an organic solvent. Addition of an organic species results in the precipitation of fission product compounds. The precipitate can be filtered from the
10 ionic liquid and calcined to convert to oxide prior to a vitrification step. Cleaned ionic liquid can then be recycled to the electrorefiner.

Combustion is another option for removal of contaminants, wherein spent ionic liquid is heated to temperatures in excess of 200°C. The disadvantage of this process
15 can be measured in economic terms: Combustion of imidazolium salts results in the production of carbon dioxide, water and a mixture of nitrogen oxides (NO_x). This means that the ionic liquid cannot be recycled and is in fact destroyed.

Since one of the most significant costs in industry will be that of the requisite raw
20 materials, any process which helps to minimise these costs through recycling is clearly a more viable option.

STATEMENT OF INVENTION

According to the present invention there is provided a method for treating a spent
25 ionic liquid composition comprising an ionic liquid and contaminant, the method comprising heating said ionic liquid to form a partial decomposition product thereof, separating said product from said contaminant and reacting the separated product with a reactant to regenerate said ionic liquid.

30 When 1-methyl-3-ethylimidazolium chloride is heated under reduced pressure, the ionic liquid partially decomposes to give 1-methylimidazole, 1-ethylimidazole,

chloromethane and chloroethane. In the initial experiments the thermolysis products were left for two days at room temperature and re-acted to produce a mixture of 1,3-dimethylimidazolium chloride, 1,3-diethylimidazolium chloride, and 1-methyl-3-ethylimidazolium chloride. These results are surprising because thermal decomposition was previously considered to destroy any potential for recycling of the ionic liquid. In the light of these experiments however thermal decomposition for the cleaning of ionic liquids is now an economically viable alternative.

Preferably the decomposition product is separated together with at least one other decomposition product from the contaminants. Separation is effected by volatilisation during the heating process. In one embodiment of the invention the partial decomposition product is separated from the at least one other decomposition product by distillation.

Preferably the ionic liquid is heated under reduced pressure at or below 2mmHg at a temperature ranging from 200°C to 300°C or more preferably from 220°C to 250°C.

DETAILED DESCRIPTION OF THE INVENTION

Spent ionic liquid, for example 1-methyl-3-ethylimidazolium chloride, is contaminated with fission products, possibly organic radiolysis products, and will require treatment in order to recover the fission products in a form suitable for conversion into a stable waste form.

In a method according to the present invention, the ionic liquid is heated under reduced pressure to a temperature within the range of 200°C to 300°C. In a preferred embodiment 1-methyl-3-ethylimidazolium is heated at a temperature from 220°C to 250°C for 2-3 hours at pressures below 2mmHg. These conditions allow for only partial decomposition of the ionic liquid and therefore the recovery of as many useful component compounds of the ionic liquid as possible. It should be understood that the conditions for partial decomposition will vary for differently substituted imidazolium salts and for different mixtures thereof.

Thermolysis of the ionic liquid results in the evolution of volatile thermolysis products. In the case of 1-methyl-3-ethylimidazolium chloride these are 1-methylimidazole, 1-ethylimidazole, chloromethane and chloroethane. Chloroethane
5 then decomposes to hydrogen chloride and ethene.

Volatile products can be collected in a cold trap and subsequent distillation of the mixture allows for separation of the components. The most volatile components are hydrogen chloride and ethene. Hydrogen chloride can be scrubbed from the system
10 using a hydroxide scrubber and ethene can either be bottled or burnt as a by-product.

1-methylimidazole and 1-ethylimidazole can be separated by distillation to allow for regeneration of the specific ionic liquid 1-methyl-3-ethylimidazolium chloride. It should be understood that separation is not required for processes which operate on a
15 mixture of 1-methyl-3-ethylimidazolium chloride, 1,3-dimethylimidazolium chloride and 1,3-diethylimidazolium chloride.

If the specific ionic liquid 1-methyl-3-ethylimidazolium chloride is required, separated 1-methylimidazole is reacted with chloroethane so as to regenerate the
20 original ionic liquid 1-methyl-3-ethylimidazolium chloride. Because some chloroethane is lost through its decomposition, fresh chloroethane is added to fully regenerate the ionic liquid. 1-ethylimidazole is reacted with chloromethane to regenerate the ionic liquid 1-methyl-3-ethylimidazolium chloride.

25 In a preferred embodiment of the invention the fission product residue is calcined to convert it into a stable waste form. In an alternative embodiment fission product residue is reacted with boric acid to convert it into a suitable form for disposal.

CLAIMS

1. A method for treating a spent ionic liquid composition comprising an ionic liquid and contaminants, the method comprising heating said ionic liquid to form a partial decomposition product thereof, separating said product from said contaminants and reacting the separated product with a reactant to regenerate said ionic liquid.
2. A method according to claim 1 wherein said partial decomposition product is separated together with at least one other decomposition product from said contaminants.
3. A method according to claim 2 or claim 3 wherein said partial decomposition product is separated from the at least one other decomposition product by distillation.
4. A method according to any of the preceding claims wherein said partial decomposition product is reacted with at least one other decomposition product produced to regenerate said ionic liquid.
5. A method according to any of the preceding claims wherein said separation is effected by volatilisation during the heating process.
6. A method according to any preceding claim wherein the ionic liquid is 1-methyl-3-ethylimidazolium chloride.
7. A method according to claim 6 wherein the partial decomposition product is a mixture of 1-methylimidazole, 1-ethylimidazole, chloromethane and chloroethane.

8. A method according to claim 7 wherein 1-methylimidazole is reacted with chloroethane to regenerate 1-methyl-3-ethylimidazolium chloride.
- 5 9. A method according to claim 5 wherein the partial decomposition product is 1-ethylimidazole.
- 10 10. A method according to claim 9 wherein 1-ethylimidazole is reacted with chloromethane to regenerate 1-methyl-3-ethylimidazolium chloride.
- 10 11. A method according to any preceding claim wherein the ionic liquid is heated under reduced pressure.
12. A method according to claim 11 wherein the pressure is at or below 2mmHg.
- 15 13. A method according to any preceding claim wherein the ionic liquid is heated to a temperature from 200 to 300°C.
14. A method according to claim 13 wherein the ionic liquid is heated to a temperature from 220 to 250°C.
- 20 15. A method according to any of claims 3 to 14 wherein volatile products resulting from volatilisation of the ionic liquid are collected in a cold trap.
- 25 16. A method according to any preceding claim wherein hydrogen chloride is produced by heating said ionic liquid and is scrubbed from the system using a hydroxide scrubber.
- 30 17. A method according to any preceding claim wherein ethene is produced by heating said ionic liquid.

18. A method according to claim 17 wherein said ethene is bottled or burnt as a by product.
- 5 19. A process for the reprocessing of nuclear fuel and the treatment of salt wastes contaminated with fission products, the process including a method of any preceding claim.
20. A process according to claim 19 wherein the fission product contaminant residue is separated and calcined before disposal.
- 10 21. A process according to claim 19 wherein the fission product contaminant residue is reacted with boric acid before disposal.
22. A method according to claim 1 and substantially as herein described.
- 15 23. Use of thermal decomposition in the treatment of spent ionic liquids wherein the decomposition products are used in the regeneration of an ionic liquid.
- 20 24. Use according to claim 23 wherein the ionic liquid is used in the reprocessing of spent nuclear fuel.

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